

## Thermodynamic properties and electrical resistivity of liquid MgZn alloys

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Received 15 November 2000, accepted 11 July 2001

**Abstract** · The observed free energy of mixing of MgZn liquid alloy is discussed on the complex formation model. The results for long-wavelength concentration-concentration fluctuations  $S_{CC}(0)$ , electrical resistivity and thermodynamic activity are presented as a function of concentration.  $S_{CC}(0)$  has been further used to obtain the chemical short range order parameter  $\alpha_1$ . Values of  $S_{CC}(0)$  and  $\alpha_1$  suggest that segregation-order transformation occurs in the MgZn liquid alloy at around 30 at % of Zn.

**Keywords** · Thermodynamic property, electrical resistivity, liquid MgZn alloys

**PACS Nos.** : 61.25.Mv, 72.15.Cz, 65.50+m

### 1. Introduction

It is fairly well known that the stability of liquid alloys is dependent on the interactions among the component atoms of the alloy [1, 2]. Liquid alloys may be considered from two extreme points of view. One is the nearly free electron model and the other is the complex forming model. Complex formation in the liquid phase is a dynamical process characterized by a finite life time [3]. It is always accompanied by a large heat of mixing, but this quantity does not always provide an accurate indication of the stoichiometric composition. Of all concentration fluctuations in the long wave length limit, the concentration-concentration fluctuation,  $S_{CC}(0)$ , has emerged as one of the most important and powerful microscopic function to understand the nature of interatomic interactions [4]. It is thought that  $S_{CC}(0)$  attains ideal values, *i.e.* random mixing of free atoms in the glass forming concentration range [5]. This glass forming range usually lies far away from stoichiometric composition [6].

Some Mg alloys are particularly interesting due to their glass forming ability and zinc is present in many commercial Mg alloys. Since the availability of Mg alloys enables the optimization of the steps necessary for light alloys production and design [7],

the study of the characteristics of alloys containing Mg is of great importance. At  $T = 923\text{K}$ , MgZn is similar in structure as CaMg and CaMg is a good glass former. MgZn glass was the first transition metal-free metallic glass [8]. It has often been observed that a good glass-forming ability is linked to a negative temperature coefficient of the electrical resistivity [9]. There exists a resistivity minima in MgZn alloy [10] at temperatures around 200-300K above the liquid temperature. MgZn is also an interesting and significant case for the study of local order and other fundamental properties. Moreover, MgZn alloy gives clear evidence of chemical short-range order (CSRO) [11-13]. The results of extended X-ray absorption fine structure measurements [8] show the occurrence of CSRO both in liquid and glassy alloys. The phase diagram of MgZn is a complicated one [14, 15]. Besides a congruently melting phase  $\text{MgZn}_2$ , there exists a number of incongruently melting compounds  $\text{Mg}_2\text{Zn}_{11}$ ,  $\text{Mg}_7\text{Zn}_3$  and  $\text{MgZn}$  and the high temperature phase  $\text{Mg}_{51}\text{Zn}_{23}$  (previously known as  $\text{Mg}_7\text{Zn}_3$ ). We have assumed the complex formation model ( $\mu A + \nu B \rightleftharpoons A_\mu B_\nu$  where  $\mu$  and  $\nu$  are integers) for the study of the thermodynamic properties and electrical resistivity of MgZn. This assumption is being used by various theoreticians [5, 6, 16-20] to investigate the alloying behaviour of such type of binary alloys.

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In this paper, the calculated results of free energy of mixing, activity,  $S_{CC}(0)$ , CSRO and electrical resistivity for MgZn liquid alloy are presented.

## 2. Complex formation model

Let a liquid binary alloy containing in all,  $N_A = C_A N$  atoms  $A$  and  $N_B = C_B N$  atoms  $B$ , be assumed to consist of  $n_1 N$  free atoms of  $A$ ,  $n_2 N$  atoms of  $B$  and  $n_3 N$  complexes  $A_\mu B_\nu$  which also act as independent scattering centers in the alloy. From the conservation of atoms, therefore,

$$\begin{aligned} n_1 &= 1 - C_B - \mu n_3, \\ n_2 &= C_B - \nu n_3, \\ n &= n_1 + n_2 + n_3. \end{aligned} \quad (1)$$

Here,  $C_A$  and  $C_B$  are the concentrations of the first and second species and  $N$  is Avogadro's number. The volume in which the free atoms are randomly distributed can be expressed as

$$\Omega = \Omega_{\text{alloy}} - n_3 N \Omega_c, \quad (2)$$

where  $\Omega_{\text{alloy}}$  is the atomic volume of the alloy and  $\Omega_c$ , the volume of the complex

$$\Omega_c = \mu \Omega_1 + \nu \Omega_2, \quad (3)$$

$\Omega_1, \Omega_2$  being the atomic volumes of the first and second species of the alloy, respectively.

The number of complexes  $n_3$  is obtained through the condition

$$\left( \frac{\partial G_M}{\partial n_3} \right)_{T,P,C} = 0, \quad (4)$$

where  $G_M$  is the Gibb's energy of mixing and  $T$  and  $P$  represent the temperature and pressure, respectively.

The conformal solution approximation [21] enables us to express  $G_M$  as

$$G_M = -n_3 g + RT \sum_{i=1}^3 n_i (\ln n_i - \ln n) + \sum_{i < j} \sum n_i n_j W_{ij} / n, \quad (5)$$

$W_{ij}$  are the interaction energies which are independent of the concentration.  $g$  is the formation energy of the complex and the term  $(-n_3 g)$  lowers the free energy of the alloy due to the formation of chemical complexes and  $R$  is the gas constant. Eqs. (4) and (5) yield

$$(n_1^\mu n_2^\nu) / (n_3^{\mu+\nu-1}) = \exp(-g/RT) \exp(y_1 + y_2 + y_3), \quad (6)$$

where

$$y_1 = (W_{12}/RT) [(\mu + \nu - 1)(n_1 n_2 / n^2) - \mu n_2 / n - \nu n_1 / n],$$

$$y_2 = (W_{13}/RT) [(\mu + \nu - 1)(n_1 n_3 / n^2) - \mu n_3 / n + n_1 / n],$$

$$y_3 = (W_{23}/RT) [(\mu + \nu - 1)(n_2 n_3 / n^2) - \nu n_3 / n + n_2 / n]. \quad (7)$$

The interaction energies  $W_{ij}$  and  $g$  are determined following the works by Bhatia and Hargrove [22]. Once the energy parameters are selected, these remain the same for all other thermodynamic investigations. Eq. (6) is solved numerically to obtain the equilibrium value of  $n_3$ , which in turn is used in (5) to evaluate  $G_M$  as a function of concentration.

## 3. Free energy of mixing and thermodynamic activity

The first step is to consider the nature of the chemical complexes ( $A_\mu B_\nu$ )  $A_\mu B_\nu$  which might exist in the liquid phase. The phase diagram [15, 23] indicates that  $\text{MgZn}_2$  is the most probable intermetallic compound. Once the values of  $\mu$  and  $\nu$  are fixed, then activity data are used to evaluate [24, 25] the values of interaction energies ( $g$  and  $W_{ij}$ ). The energy parameters evaluated for  $\text{MgZn}$  are

$$g/RT = +2.65, W_{12}/RT = 1 - 0.004 - 0.04, W_{13}/RT = 0.15,$$

$$W_{23}/RT = -0.50.$$

In the light of the complex formation energy,  $\text{MgZn}$  is a weakly interacting system [2, 6, 22, 24] like  $\text{AlCa}$ ,  $\text{AlMg}$ ,  $\text{MgSn}$ ,  $\text{AgAl}$  and  $\text{CuSn}$ . Eq. (6) has been used to obtain equilibrium values of  $n_3$  and hence  $G_M$  via eq. (5). The  $n_3$ - $C$  and  $G_M$ - $C$  curves for  $\text{MgZn}$  are displayed in the upper and lower parts of Figure 1 respectively. The maximum value of  $n_3$  occurs at  $C_{Zn} = 0.66$ . The computed values and observed values of  $G_M$  [23] are in reasonable agreement. It may be observed that the variation of free energy is not symmetrical about  $C = 1/2$

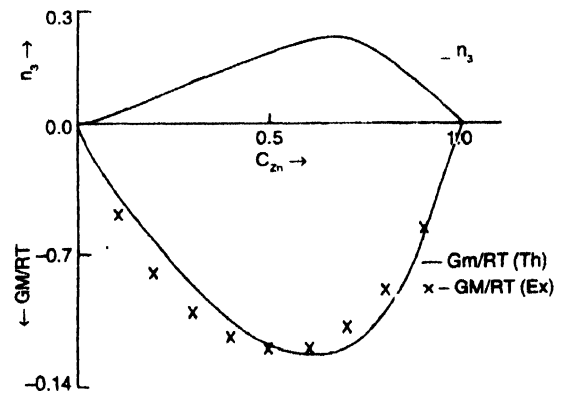


Figure 1. Upper side : Number of complexes ( $n_3$ ) versus concentration of Zn ( $C_{Zn}$ ). Lower side : Free energy of mixing ( $G_M/RT$ ) versus concentration of Zn ( $C_{Zn}$ ).

Eq. (5) for the free energy of mixing can also be used to obtain an expression for the activity  $a_A$  for the atom type  $A$  as follows

$$\begin{aligned}
 RT \ln a_A &= (\partial G_M / \partial N_A)_{T,P,N} \\
 &= G_M + (1 - C_A) (\partial G_M / \partial C_A)_{T,P,N} \quad (A = \text{Mg}), \quad (8) \\
 &\quad (B = \text{Zn}).
 \end{aligned}$$

Eqs. (5) and (8) yield

$$\begin{aligned}
 \ln a_A &= 1 - n + \ln(n_1 / n_2) + \ln n + n_3 (W_{13} / RT) \\
 &+ n_2 (W_{12} / RT) - \sum_{i < j} n_i n_j W_{ij} / RT. \quad (9)
 \end{aligned}$$

$\ln a_{\text{Mg}}$  has been computed through (9) as a function of concentration. The computed values of activity of Mg in MgZn alloy are given in Table 1 along with the experimental observations [23]. Table 1 shows that theoretical and experimental values are in reasonable agreement.

Table 1. Thermodynamic activity  $a_{\text{Mg}}$  of MgZn liquid alloys at 923K.

$C_{\text{Mg}}$	Calculated	Observed [23]
0.1	0.007	0.015
0.2	0.031	0.053
0.3	0.119	0.121
0.4	0.281	0.217
0.5	0.452	0.336
0.6	0.604	0.473
0.7	0.731	0.613
0.8	0.836	0.747
0.9	0.922	0.866

#### 4. Long-wavelength concentration correlation function and chemical short-range order

The long-wavelength limit of the three correlation functions, namely the concentration-concentration correlation functions  $S_{CC}(0)$ , the number-number correlation functions  $S_{NN}(0)$  and the number-concentration correlation functions  $S_{NC}(0)$ , have been widely used to understand the stability [5, 6, 25, 26] of the binary mixture. The experimental values of  $S_{CC}(0)$  can readily be derived from the thermodynamic functions.

$$\begin{aligned}
 S_{CC}(0) &= RT (\partial^2 G_M / \partial C_A^2)_{T,P,N} \\
 &= (1 - C_A) a_A (\partial a_A / \partial C_A)^{-1}_{T,P,N} \\
 &= C_A a_B (\partial a_B / \partial (1 - C_A))^{-1}_{T,P,N}. \quad (10)
 \end{aligned}$$

Obviously  $S_{CC}(0)$  can be obtained directly from the observed activity data following the last two equalities of (10) or may be obtained theoretically with the help of (5) and (10)

$$S_{CC}(0) = \left[ \sum_{i=1}^3 n_i'^2 / n_i - n'^2 / n + (2n / RT) \sum_{i < j} n_i' n_j' W_{ij} / n^2 \right]^{-1} \quad (11)$$

where a prime denotes differentiation with respect to  $C_A$ ,

$$n' = n_1' + n_2' + n_3',$$

$$n_1' = 1 - \mu n_3',$$

$$n_2' = -1 - \nu n_3',$$

$$n_3' = P / Q,$$

$$\begin{aligned}
 P &= \mu / n_1 - \nu / n_2 - (\mu + \nu - 1)(1 / n^2) ((n_2 - n_1) W_{12} / RT \\
 &+ (n_3 / RT) (W_{13} - W_{12})) - ((\mu + \nu) W_{12} + W_{13} - W_{23}) (1 / nRT),
 \end{aligned}$$

$$Q = ((\mu + \nu - 1)(y_1 + y_2 + y_3) + \mu \nu W_{12} - \mu \cdot W_{13} - \nu \cdot W_{23}).$$

$$(2 / nRT) + \mu^2 / n_1 + \nu^2 / n_2 + 1 / n_3 - (\mu + \nu - 1)^2 / n. \quad (12)$$

The values of  $S_{CC}(0)$  computed for MgZn liquid alloy following eq. (11) are shown in Figure 2. These are compared with the values obtained directly from the observed activity data [23] using the last two identities in (10) and with those computed by Ramchandra Rao *et al* [26]. It is clear from Figure 2 that the values of  $S_{CC}(0)$  obtained from activity data and those by Rao *et al* are always lower than  $S_{CC}^{id}(0)$  ( $S_{CC}^{id}(0) = C_A C_B$ ). In contradiction of these two values, our computed values of  $S_{CC}(0)$  are higher than  $S_{CC}^{id}(0)$  upto  $C_{\text{Zn}} < 0.28$ . For  $C_{\text{Zn}} \geq 0.28$ ,  $S_{CC}(0)$  is smaller than  $S_{CC}^{id}(0)$ . The segregation (like atoms pairing) is expected upto the region  $C_{\text{Zn}} < 0.28$  since  $S_{CC}(0) > S_{CC}^{id}(0)$ . After  $C_{\text{Zn}} = 0.28$ , computed values of  $S_{CC}(0) < S_{CC}^{id}(0)$  from which existence of chemical ordering leading to complex formation (unlike atoms pairing) is expected. It may be seen from Figure 1 that the fraction of chemical complexes  $n_3$  is a maximum at  $C_{\text{Zn}} = 0.66$  but a shoulder in  $S_{CC}(0)$  in Figure 2,

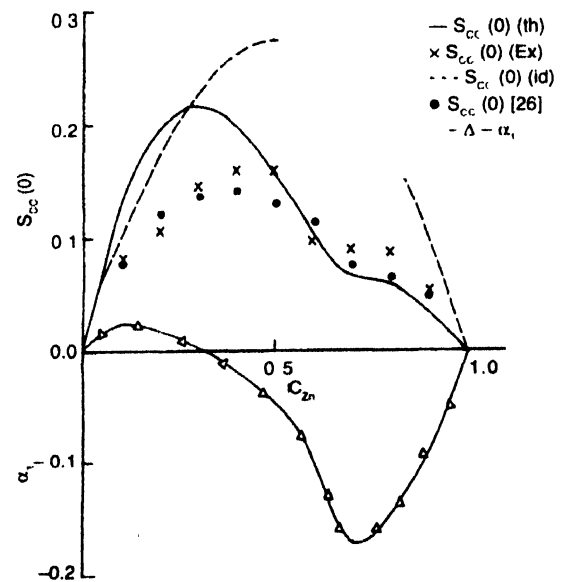


Figure 2. Concentration fluctuations in the long-wavelength limit ( $S_{CC}(0)$ ) and chemical short-range order ( $\alpha_1$ ) versus concentration of Zn ( $C_{\text{Zn}}$ ).

slightly shifts towards larger Zn content at  $C_{Zn} = 0.75$ . It is because the position of the minimum in  $S_{CC}(0)$  depends collectively on  $\mu, v, g$  and  $W_{ij}$ .

The non-resemblance of computed values of  $S_{CC}(0)$  with those obtained directly from activity data, may be attributed to the inaccuracies involved in the numerical differentiation of the activity data, particularly at the two ends of  $C_{Zn}$ . The activity of one of the components becomes negligibly small at the either end of the  $C_{Zn}$  scale, which creates problems in the numerical differentiation.

The Warren-Cowley [27, 28] short-range order parameter  $\alpha_1$  for MgZn liquid alloy has been computed to quantify the degree of order.  $\alpha_1$  can be determined experimentally for  $S_{NN}(q)$  and  $S_{CC}(q)$  but it is difficult to determine these parameters for all kinds of binary alloys [25, 29, 30].  $\alpha_1$  can be obtained theoretically [5, 25] as

$$\alpha_1 = (S - 1) / (S(Z - 1) + 1), \quad (13)$$

$$S = S_{CC}(0) / S_{CC}^{id}(0), \quad (14)$$

$Z$  is the coordination number, which is taken as 12 for our purpose. The computed values of  $\alpha_1$  computed through (13) has been shown in Figure 2. The positive value of  $\alpha_1$  ( $\alpha_1 > 0$ ) upto  $C_{Zn} < 0.3$  refers to like atoms pairing and negative value of  $\alpha_1$  ( $\alpha_1 < 0$ ) for  $C_{Zn} > 0.3$  refers to unlike atoms pairing as nearest neighbours. The minimum value of  $\alpha_1$  occurs near  $C_{Zn} = 0.7$ . This indicates that Zn-rich end is more ordered than Mg-rich end.

## 5. Electrical resistivity

We can express the electrical resistivity of a ternary mixture as [18]

$$R = (12\pi \cdot \Omega_s) / (\hbar e^2 \cdot V_F^2) \int_0^1 d(q/2k_F) (q/2k_F)^3 \sum_{i,j=1,2,3} (X_i V_i^2(q) S_{ii}(q) + 2(X_i X_j)^{1/2} V_i(q) V_j(q) S_{ij}(q)). \quad (15)$$

Here,  $V_F$  is the Fermi velocity,  $\Omega_s = \Omega / n$  the scattering volume of the alloy in which free electrons travel,  $q$  the phonon wave vector,  $V_i(q)$  ( $i = 1, 2$ )  $= W_i(q) / \epsilon^*(q)$  are the screened form factors of Mg, Zn and  $V_3(q)$  is the screened form factor of the complex MgZn<sub>2</sub>.  $X_i = n_i / n$  etc are the concentration functions of different scattering centers.  $k_F$  is the Fermi wave vector.

In order to compute form factors of Mg and Zn, the Heine-Abarenkov model potential

$$W_i(q) = \frac{4\pi Z_i}{q^2 \Omega} \cos(q r_m) - \frac{4\pi A_0}{\Omega q^3} (\sin(q r_m) - q r_m \cos(q r_m)) \quad (16)$$

has been considered. Here  $r_m$  is the model radius,  $A_0$  is the well depth.  $\epsilon^*(q)$  is the modified Hertz dielectric function given by

$$\epsilon^*(q) = 1 + (\epsilon(q) - 1)(1 - G(q)). \quad (17)$$

The Vashishta and Singwi [31] form of exchange  $G(q)$  has been used for correlated motion of the conduction electrons.

$S_{ij}(q)$  are the partial structure factors which have been obtained following the work of Hoshino [32]. The hard sphere diameters  $\sigma_1, \sigma_2$  occur as the important ingradient. Those  $\sigma_1, \sigma_2$  have been taken which reproduce the first peak of structure factor of pure liquid Mg and Zn at their respective melting points. These diameters are taken as concentration-independent. The hard sphere diameter of the complex  $\sigma_3$  is taken as a parameter [18]. The scattering potential  $V_3(q)$  for the complex has been estimated as [18]

$$V_3(q) = \frac{\mu V_1(q) + v V_2(q)}{\mu + v}, \quad (18)$$

since  $V_3(q)$  can not be obtained from first principle as the complex has flexible structure.

The resistivity of MgZn liquid alloy calculated through (15) has been depicted in Figure 3 along with the experimental observation by Walhout *et al* [33]. There is a fair agreement between the theoretical and experimental values except for pure

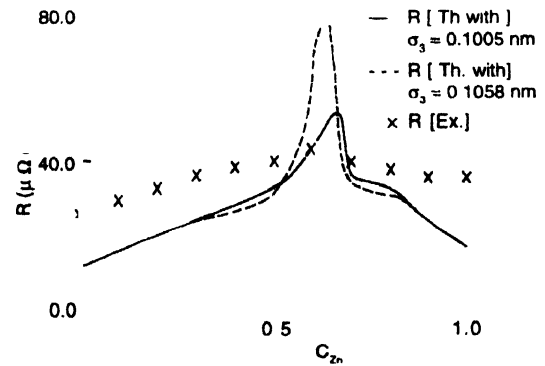


Figure 3. Electrical resistivity ( $R$ ) versus concentration of Zn ( $C_{Zn}$ )

Mg and Zn. To see the effect of  $\sigma_3$ , the two values of resistivity were calculated with  $\sigma_3 = 0.1005$  nm and  $\sigma_3 = 0.1058$  nm and the results are shown in Figure 3. It is found that the rise in the value of  $\sigma_3$  not only increases the electrical resistivity of MgZn liquid alloy, but the maximum shifts towards Mg rich end also. The maximum value of resistivity comes at stoichiometric composition. The difference between observed and theoretical resistivity of pure Mg and Zn may be due to the fact that we

have not taken into consideration the non-local contribution in the calculation of form factors. Also, Zn is the metal which gives most trouble due to *d*-bonding. There is also a large mismatch at intermediate concentration. Specially, the experimental curve shows no maximum as in the case of theoretical curve, but only a slight hump. By considering the presence of another complex  $\text{Mg}_2\text{Zn}_3$  along with  $\text{MgZn}_2$  [14] in the intermetallic phase and a phase shift expansion of form factor  $V(q)$  for Zn [34], the mismatch between the computed and experimental values may be minimized.

## 6. Conclusion

- (a) In the concentration range  $0 \leq \text{Zn} \leq 0.28$ , the like *i.e.*, Mg-Mg and Zn-Zn atoms are pairing as nearest neighbours.
- (b) Both  $S_{CC}(0)$  and  $\alpha_1$  suggest that segregation-order transformation occurs around 30 at % of Zn. Thus, MgZn liquid alloy is a glass former for a certain range of composition.
- (c)  $S_{CC}(0)$  exhibits only a weak deepening around the concentration  $\text{MgZn}_2$ . Here small values of the size effect 1.52 and electro negativity 0.0392 are not responsible for producing asymmetry in  $S_{CC}(0)$  and  $\alpha_1$ .
- (d) It is also observed from the low value of electro negativity *i.e.*, 0.0392 as compared with *e.g.* 0.51 for CsAu, that MgZn possesses weaker ionic character.
- (e) The hard sphere diameter of the complex plays an important role in the calculation of partial structure factors and thereby inelectrical resistivity.

## Acknowledgment

The financial support by the Council of Scientific & Industrial Research, New Delhi is gratefully acknowledged by A. K. Mishra. The authors are thankful to Dr. R. N. Singh, Prof. of Physics, S. Q. University, Oman for encouragement and Dr. B. B. Sahay, Retd. Prof. of Physics, B. C. E., Bhagalpur for useful discussions.

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